IGNITION AND INCENDIVITY OF SINGLE MICRON SIZE MAGNESIUM PARTICLES IRRADIATED BY A LASER BEAM

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INTRODUCTION

This paper describes an experimental study of the ignition and incendivity characteristics of magnesium particles heated by intense radiation. While many investigations have been concerned with determining ignition temperatures of metal particles, much of their work has been with the ignition of single particles by hot gases.(1,2,3)* In dust or hybrid flames, however, a large portion of the particle ignition energy is supplied through radiation. Indeed, in a study of dust flames of aluminum and graphite,(4) the contribution of radiation was estimated to be 30 to 60 percent of the total ignition energy. The ignition of combustible gases by flying abrasion sparks and the incendivity of heated passive pellets shot into explosive gases have been examined by many investigators(5,6,7); however, the relative motion between particles and environment complicates the analysis and interpretation. In the present study, single micron size magnesium particles were radiatively heated while suspended in quiescent cold environments of oxygen, air, He-oxygen, and methane-air mixtures, and the ignition and incendive mechanisms of the particles were examined.

EXPERIMENTAL APPARATUS AND PROCEDURE

Figure 1 shows a schematic of the experimental apparatus. The laser used a 1.3-cm-diameter neodymium-doped glass rod and had a pulse duration of about 0.9 millisecond. The laser beam was collimated by a simple 17-cm-focal-length convex lens positioned 9.5 cm from the magnesium particle suspended in the center of the levitation device. Devices of this nature have been used by other investigators.(8) The levitator consisted of four 0.3-cm-diameter by 8-cm-long vertically mounted metal rods which formed a 1.4-cm square. The rod ends were supported by two teflon insulating disks; alternate rods were connected together and to a 200- to 1500-volt AC supply. Two metal circular plates attached to the inner face of the top and bottom teflon disks and insulated from the rods were connected to a DC source of 100 to 1000 volts. In practice several milligrams of the magnesium dust placed in a centrally located cavity in the bottom metal disk were dispersed in the apparatus by charging with a high-voltage pulse. A few of the scattered particles were captured in the levitation apparatus and retained along the central vertical axis of the device by the rotating electric field.

All particles but one were readily eliminated by varying the AC and DC potentials; the single remaining particle was then positioned in line with the laser beam and its size determined by use of a microscope. The levitator was contained in a transparent glass chamber 6 x 6 x 10 cm having two vents for use with flammable mixtures. In the experiments, single spherical magnesium particles (99 percent pure) ranging in size from 28- to 120-microns diameter were suspended and irradiated in the glass chamber after filling with dry gas mixtures of air, helium-20 percent oxygen, pure oxygen, pure argon and stoichiometric methane-air. Streak and high-speed framing cameras were used to photograph the particles during irradiation. In order to determine when particle ignition occurred, a sample of the laser beam was picked up by the photodiode (fig. 1) and amplified to energize

* Underlined numbers in parentheses refer to references at the end of the paper.

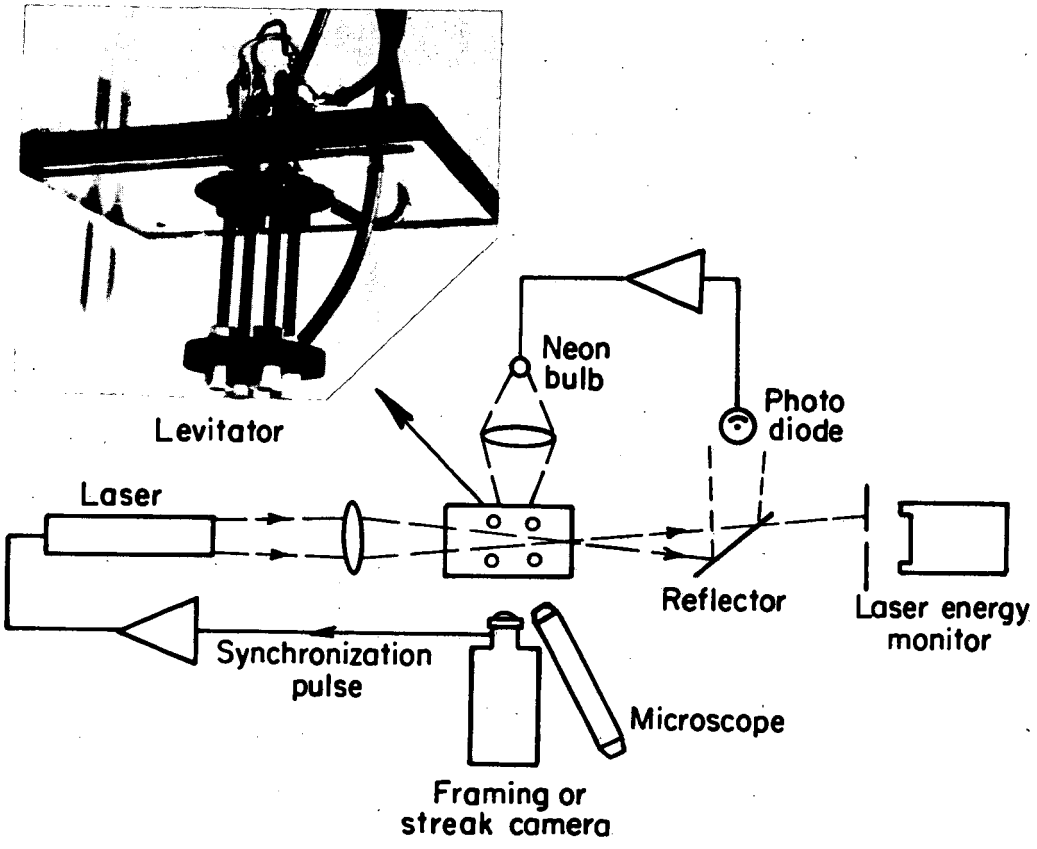

Fig. 1. - Apparatus for laser ignition of single magnesium particles.

a neon bulb whose focused image on the camera film correlated with the laser pulse. The laser pulse duration was measured by connecting the photodiode output to an oscilloscope. Due to the characteristic energy inhomogeneity in the laser beam cross-section, the magnesium particle was reproducibly positioned in a small, relatively uniform section of the laser beam and a 1-mm-diameter aperture in front of the laser energy monitor limited the measured energy to this uniform section. The monitor was a radiometer based on the calorimetric principle and measured total pulse energy passing through the apparatus. The collimating lens position was fixed and laser beam energy was varied by changing the charge voltage on the laser flash tube capacitor bank. Since the magnesium particle blocked a small portion of the laser beam, the laser energy was monitored between tests in the absence of a particle. The calculated laser power densities irradiating the particle were a function of the measured laser energy passing through the aperture, the aperture cross-section projected back to the particle site, and the pulse duration.

Schlieren photographs were taken during the irradiation of single magnesium particles (60- and 120-micron diameter) suspended in air, pure argon and stoichiometric methane-air mixtures. In the experimental arrangement shown in figure 2, a ruby laser having an approximate 0.9 millisecond length pulse was convenient to use for the radiation heating source. It was also advantageous to use the illustrated glass light guide to reflect a portion of the laser light on the film for time correlation of the irradiated particle with the laser pulse.

RESULTS AND DISCUSSIONS

Particle Ignition

In previous hot-gas ignition experiments of single magnesium particle in air, it was established that gas temperatures of 640° to 740°C are necessary for ignition of particles ranging in diameter from 120 to 20 microns, respectively(3); the observed gas temperature increase with decreasing particle size was attributed to the increased heat loss per particle surface area with decreasing particle diameter during the ignition process. In our experiments, the particle was initially surrounded by a cold gas; therefore, higher particle temperatures were necessary for ignition to compensate for the increased heat losses to the cold ambient atmosphere. In addition, maximum temperatures attained by a particle during irradiation by a short pulse depend on the particle heat capacity, thus requiring that the ignition energy supplied by the laser increase with particle size.

Figure 3 is a streak photograph of a laser-ignited, 84-micron-diameter magnesium particle burning in air. Particle spiraling during combustion, shown in this photograph, was frequently observed and is ascribed to preferential burning of the particle. In many cases the particle was seen to fragmentize following ignition. Burning lifetimes of the particles were usually much shorter than reported in the previous hot-gas ignition studies.

Figure 4 is a plot of our experimental data showing the laser beam power density required to ignite single magnesium particles suspended in various atmospheres. At these critical powers, ignition always occurred near the end of the laser pulse. As laser power was increased above the critical value, ignition delay time could be shortened appreciably. As expected, the radiant power for ignition increases with particle size and with increased thermal conductivity of the ambient gases (thermal conductivity of helium is approximately 5 times that of air). The similar critical radiant powers required for particle ignition for air and pure oxygen agree qualitatively with results of the hot-gas ignition experiments (1) in which ignition temperature was found to be approximately equal for magnesium particles in air and pure oxygen; these findings suggest that oxygen diffusion is not a controlling factor in the ignition mechanism.

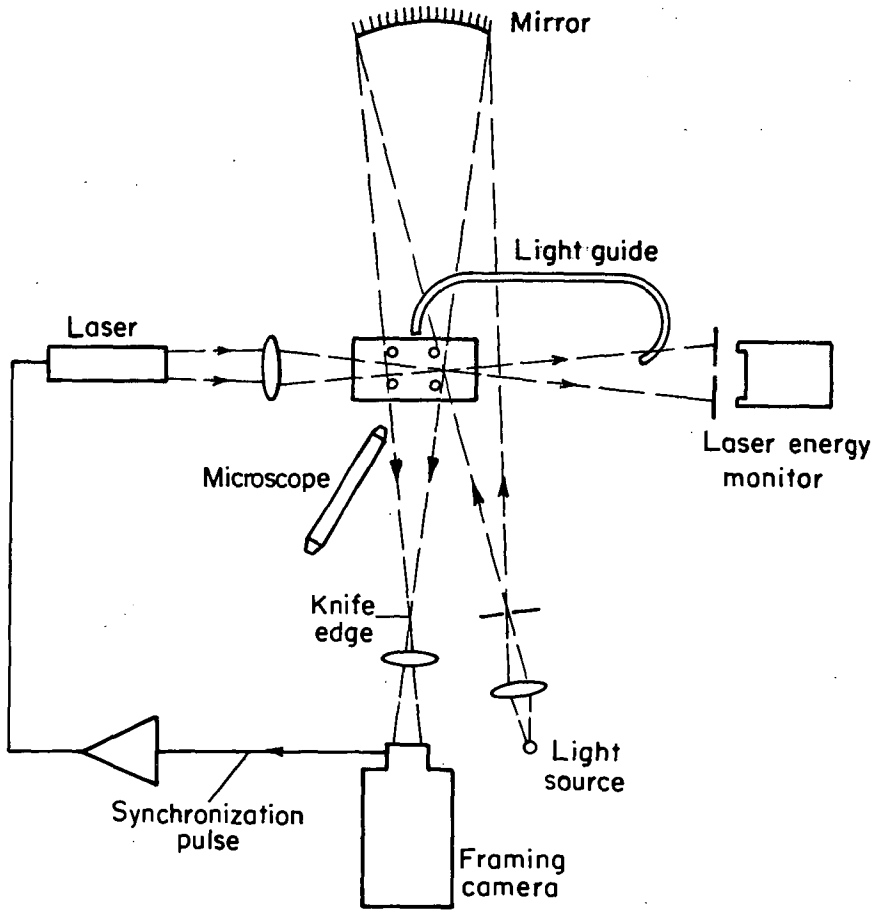

Fig. 2. - Apparatus for schlieren study.

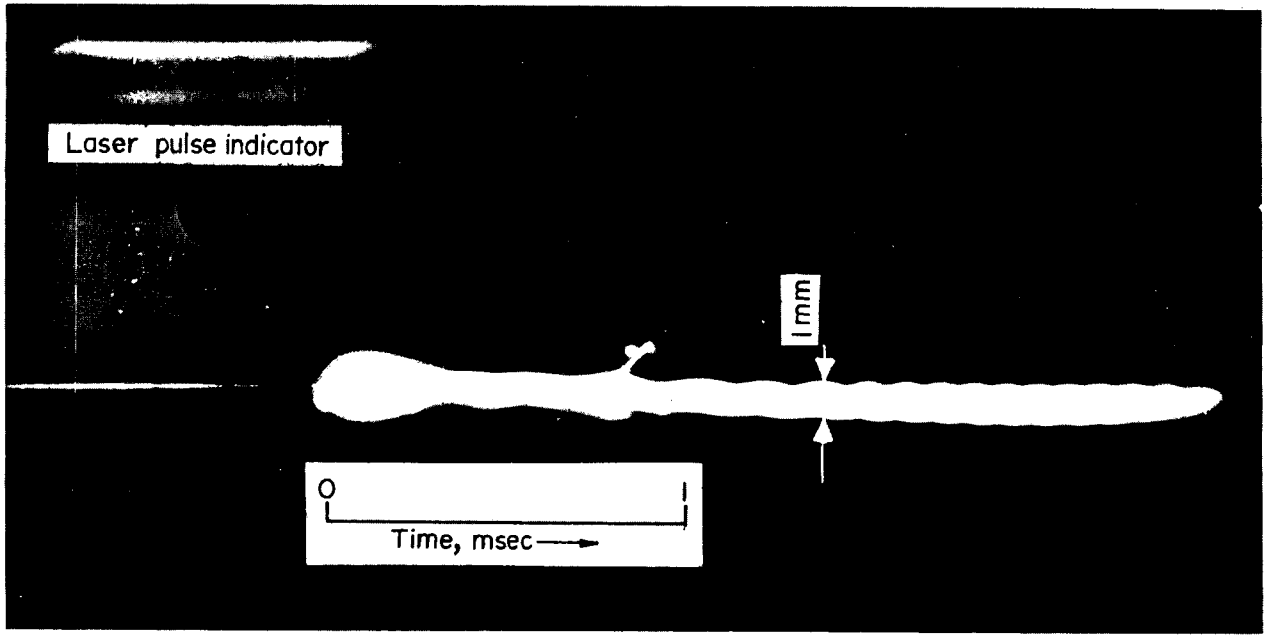

Fig. 3. - Streak photograph of laser ignited 84 micron diameter particle burning in air.

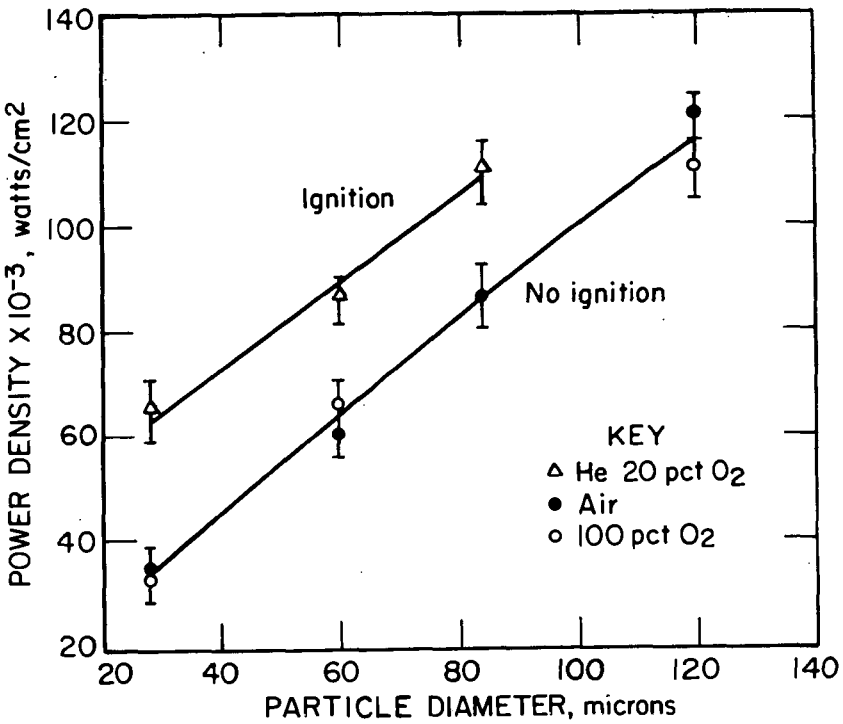

Fig. 4. - Laser power density required to ignite single magnesium particles in various atmospheres.

In additional experiments, single particles (60- and 84-micron diameter) suspended in pure argon and illuminated by a long-duration photoflash light were irradiated by laser powers equal to that previously required for ignition in air. The photoflash light revealed the rapid growth of a cloud surrounding the particle (presumably, condensed metal vapors) near the termination of the laser pulse. The particle was usually still discernable within the cloud.

A sequence of schlieren photographs of a 60-micron-diameter particle irradiated in pure argon is shown in figure 5; the laser power density was again just sufficient to ignite a similar size particle in air. The expanding schlieren image is considered to be mainly due to the expelled hot metal vapor, and is first visible near the mid-point of the laser pulse. A graph of the schlieren growth rate for an irradiated particle suspended in argon and in air is shown in figure 6. The increased growth of the schlieren taken in air is indicative of the magnesium-air reaction process.

In previous investigations of magnesium particle ignition by hot gas, the maximum required gas temperature was 740°C and in the ignition of magnesium ribbon by resistance heating,(9) metal temperatures were estimated to be in the vicinity of the metal melting point (621°C) during the onset of ignition. Our experiments suggest that ignition of a magnesium particle by a 0.9 msec intense radiation pulse occurs in the vapor phase. During the radiation pulse the particle's surface rises to the boiling point, the particle vaporizes, and ignition follows. For critical radiant powers, vaporization begins near the mid-point of the laser pulse and ignition occurs in the proximity of the pulse end.

Table I shows the average burning lifetimes of magnesium particles ignited by critical laser energies as compared to hot-gas ignition experiments. The shorter lifetimes for the laser-induced ignitions are ascribed to particle vaporization prior to ignition and disintegration of the particle during combustion.

TABLE I

Average burning lifetime of laser-ignited magnesium particles in air compared with hot-gas ignition

Diameter, microns	Lifetime, msec	
	Laser ignited	Hot gas ignited
120	5.0	16.8
84	2.4	6.2 (80 micron)
60	.9	1.8 (50 micron)
28	.1	

For the relatively low laser radiant power densities used in our experiments and large thermal diffusivity of the particle, the particle has a relatively small temperature gradient. A rough approximation of this gradient can be obtained from the relationship $\epsilon E_0 = \lambda dT/dr$, where ϵ is the surface coefficient of absorptivity (assumed to have a value of 0.3), E_0 is the laser beam power density, λ is the metal thermal conductivity having an average value of 1 Joule/cm sec °C, T is the temperature, and r is the particle radius. From this relationship the temperature difference between the particle surface and center would be of the order of 9 to 180°C for the 28 to 120 micron particle respectively using the data of figure 4 for the air environment. For a radiantly heated magnesium particle suspended in an inert atmosphere, the time for the particle surface to begin boiling and the quantity of metal vaporized during the pulse can be readily calculated if the following assumptions are made.

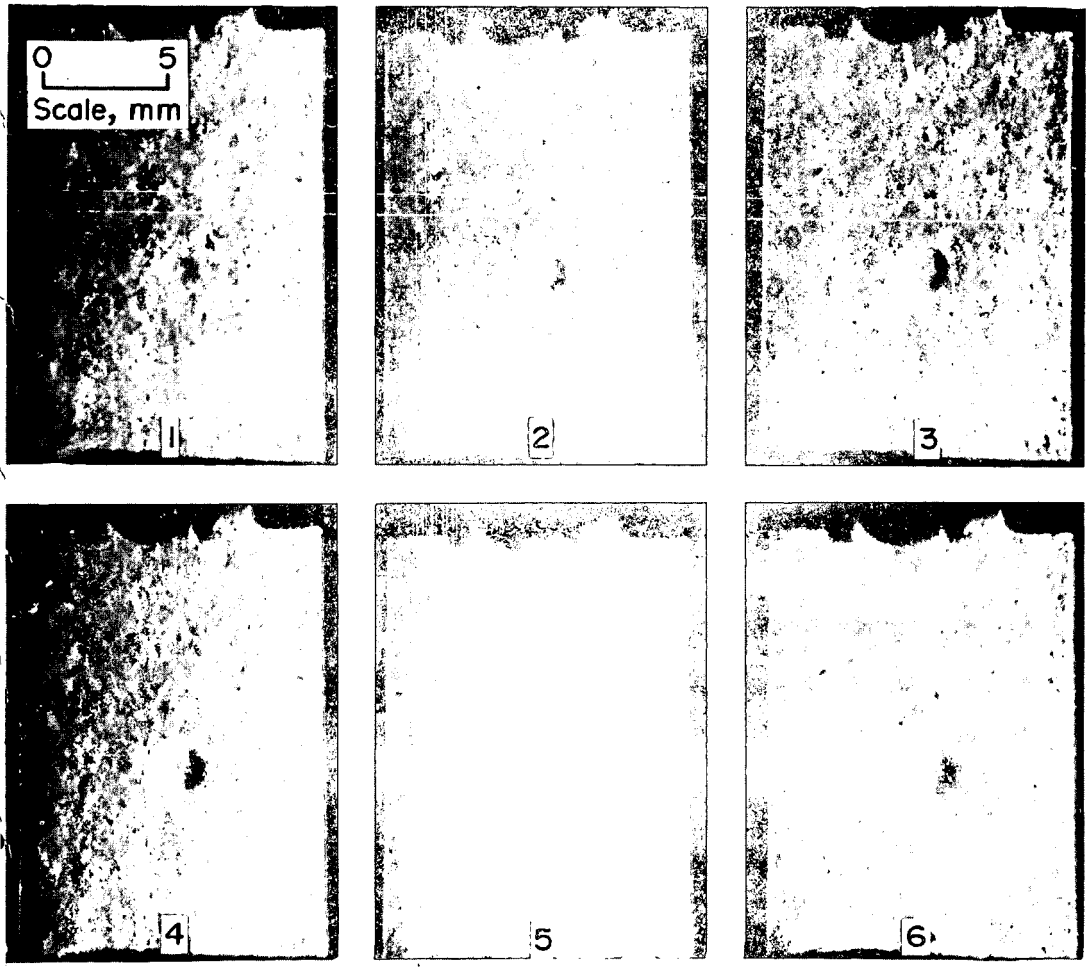

Fig. 5. - Schlieren records of a laser irradiated 60 micron particle in pure argon, 0.13 msec between frames.

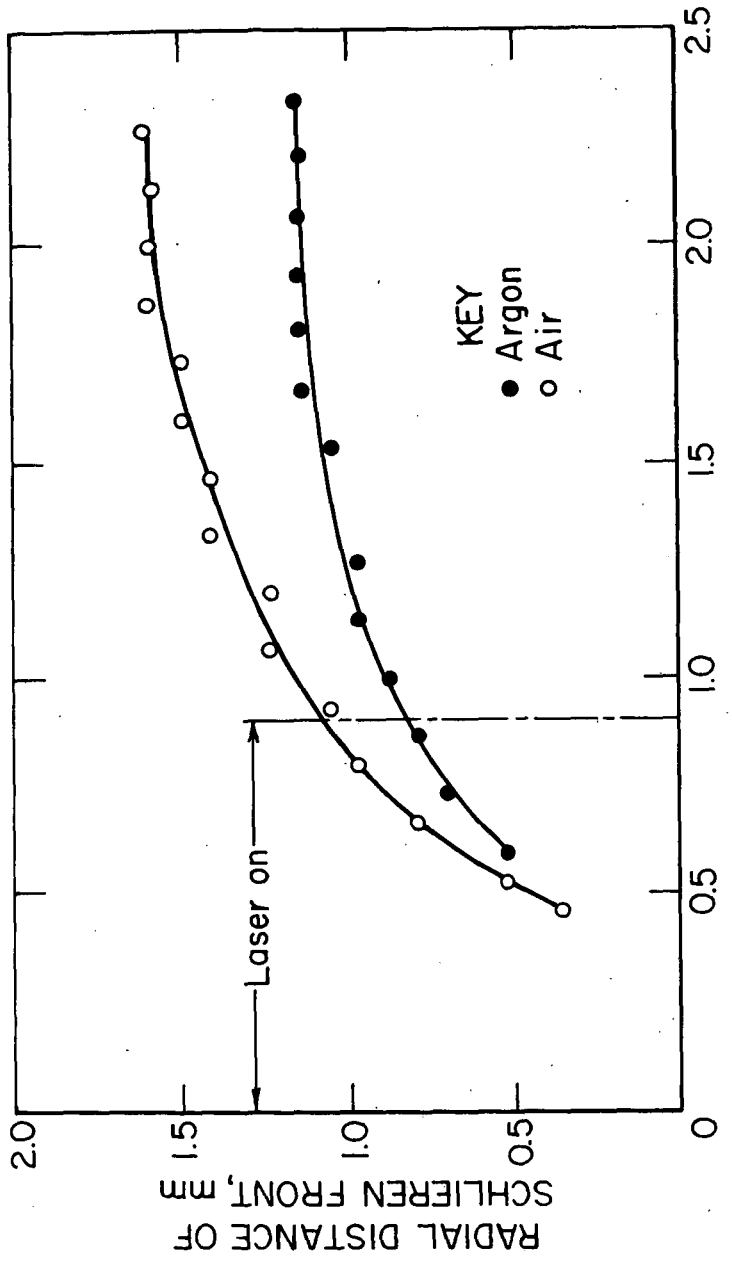

Fig. 6. - Growth of schlieren sphere surrounding a laser irradiated 60 micron diameter magnesium particle.

(1) The radiant flux absorbed by the particle is equally distributed over the entire particle surface. The particle surface coefficient of absorptivity (ϵ) is given an average value of 0.3 -- the thickness of oxide coating on the particle is less than several hundred Angstroms and has little effect on ϵ or the heat conduction.

(2) The temperature distribution within the particle is uniform.

(3) Radiative heat losses and heat transfer to the surrounding gases can be neglected.

Using these assumptions, the energy balance equation for the particle can then be written:

$$\epsilon E_o \pi r^2 t_b = \frac{4}{3}\pi r^3 \rho \left[C_\rho \Delta T + H_M\right], \quad (1)$$

where E_o is the radiant power density in joules/cm^{2}sec, r is the particle radius in cm, t_b is the time in seconds for the particle to reach the boiling point, ρ the average particle density = 1.7 gms/cm^3, C_p the average particle specific heat = 1.3 joules/cm ^oC, ΔT the temperature difference from ambient to the metal's boiling point (1100^oC), and H_M is the heat of fusion of the metal (380 joules/gm). The reduction in particle size due to evaporation can be expressed by:

$$E_o \pi r^2 = L_v \rho 4 \pi r^2 \frac{dr}{dt} \quad (2)$$

where L_v is the heat of vaporization (5500 joules/gm) and t is the time remaining in the laser pulse after the particle surface reaches the boiling point.

Using these equations, table II shows the calculated times for particles to reach the boiling point and the final particle size for critical laser powers obtained from figure 4 for air environments.

TABLE II

Boiling times and final particle size for laser irradiation at critical power densities

Particle diameter, microns	Time to boiling point, msec	Final particle diameter, microns
28	0.52	26
60	.62	56
84	.65	80
120	.70	116

The calculated times to reach the boiling point for the small particles (table II) are approximately 20 percent larger than our experimental results; the increased calculated values for the larger particles are probably due to our assumption of a uniform temperature within the particle. The small reduction in particle size due to vaporization prior to ignition accounts only partially for the short burning lifetimes noted in our experiments. In addition, the possible absorption of laser energy by the vaporized metal would be effective in reducing the particle absorptivity and thereby lessen the vaporization rate. Results of this analysis suggest that particle disintegration is the most significant factor in decreasing the burning lifetimes.

Incendivity

Figure 7 shows a plot of the critical radiant power density required for the incendive ignition of stoichiometric methane-air mixtures by irradiatively heated single magnesium particles. For comparison purposes, the curve of particle ignition in air from figure 4 is also included. It is significant to note that the 120-micron-diameter particle requires less radiant power for incendive ignition than for particle ignition in air, whereas critical radiant power for the 84- and 60-micron particles appear to be similar to the values obtained in air, and the smaller 50- and 28-micron particles need greater radiant power for incendivity than for particle ignition. This latter observation indicates that the smaller particles can be burned in a flammable environment without igniting the environment.

Table III lists the energy released during combustion of magnesium particles and the computed radiant energy absorbed by the particles during the laser pulse using equations 1 and 2 and the critical radiant powers for incendivity shown in figure 7. Information for a 20-micron-diameter particle, which did not ignite the methane-air mixture when irradiated with radiant power densities as high as 270 watts/cm^2, is also listed in this table for comparison purposes.

TABLE III

Energy released during combustion and radiant energy absorbed by magnesium particles during irradiation

Particle diameter, microns	Energy of combustion, millijoules	Energy absorption, millijoules
120	36.7	2.7
84	12.6	1.3
60	4.6	.5
50	2.7	.4
28	.5	.1
20	.16	.05

All the particles listed in table III except for the 28 and 20 micron, have greater absorbed energies than the reported (0.25 mJ) minimum spark ignition energy for stoichiometric methane-air mixtures. However, gas ignition by relatively slowly heated hot bodies should require considerably more energy than by short duration sparks. Nevertheless, since the 120-micron particle requires much less radiant flux for incendive ignition than for particle ignition in air, then this particle apparently ignites the methane-air mixture by hot-body mechanisms. Therefore, neglecting any magnesium-O_2 reaction, the minimum ignition energy of the gas mixture by small hot particles (1100°C) should be of the order of 3 mJ. The smaller particles apparently ignite first then in turn ignite the gas mixture since the energy required for incendive ignition is as great or greater than that required for particle ignition. The drop in the radiant power density required for incendivity ignition for the 60-micron particle shown in figure 7 is evidently due to an interaction of particle ignition and methane-air ignition processes. The 50- and 28-micron particles require energies in excess of particle ignition in order to shorten their burning lifetimes and thereby enhance the incendive process. The combustion and absorbed energy calculated for the 20-micron particle is seen to be less than the spark ignition requirements, therefore nonignitability by this particle size is not surprising. Rae(7) estimated the minimum mass of rapidly moving and burning magnesium particles capable of igniting a methane-air mixture to be 1.0 microgram which corresponds to a 105-micron-diameter sphere. This value is almost fourfold greater than the minimum particle size used in our experiments and illustrates the probable effect

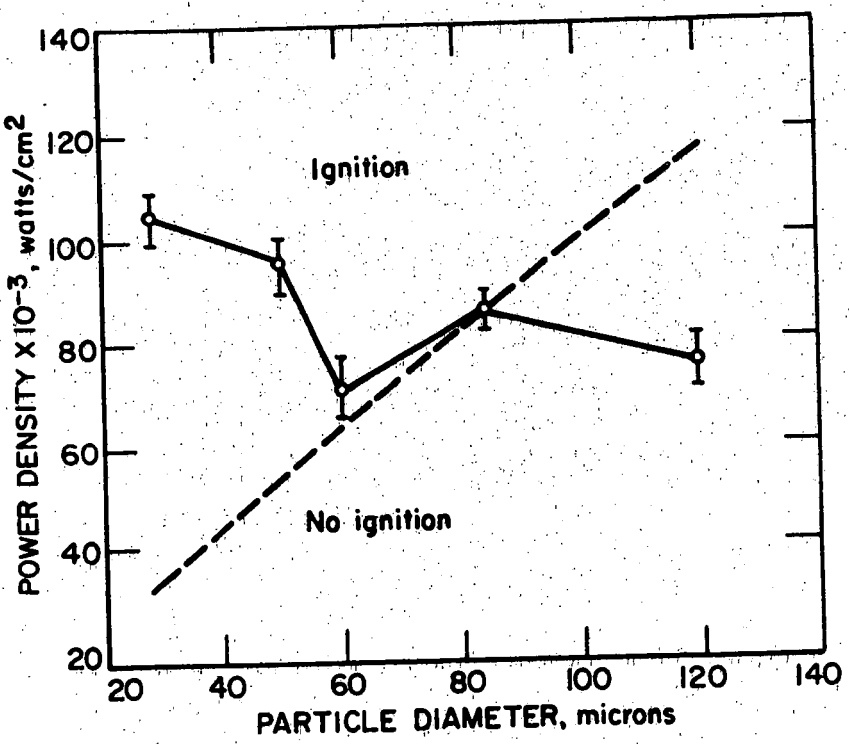

Fig. 7. - Laser power density required for single magnesium particle to ignite stoichiometric methane-air mixture.

of particle motion and burning lifetime on incendive efficiency. The effect of particle size and motion on hot-body ignition is demonstrated in work by Silver(5) and Paterson(6) who ignited gas mixtures with heated platinum and quartz spheres; their results showed that ignition temperature of the spheres increased with decreasing sphere diameter and increasing sphere velocity through the fuel mixture.

Figure 8 shows a sequence of schlieren photographs obtained during the irradiation of a 60-micron-diameter magnesium particle suspended in a stoichiometric methane-air mixture. Figure 9 is a plot of the growth of the schlieren image during the irradiation of a single 60-micron and a 120-micron-diameter particle using the radiant power density (critical) necessary for incendive ignition of stoichiometric methane-air mixtures. Figure 10 is a similar plot for a 120-micron particle irradiated by critical and twice-critical radiant power densities. The results of these studies suggest that the incendive mechanism can be described as a three-stage process; the first stage being an extremely rapid growth of a spherical envelope surrounding the particle resulting from the expanding hot metal vapors and magnesium-methane-air reaction; the second stage is characterized by a much slower growth rate and is considered to result from the continued pre-ignition process of the methane-air reaction; the third stage is the methane-air combustion front and corresponds to an expanding front traveling at a constant velocity. The rate of growth of all stages is observed to depend on laser energy and particle size. Stoichiometric methane-air flame speeds should be about 270 cm/sec. This corresponds approximately to the value obtained for the 120-micron particle irradiated by twice critical power shown in figure 10. Hot-wire ignition experiments of methane-air systems by Ashman(10) showed a similar pattern, a decreased flame speed with a decrease in wire temperature. His results also indicated that an exothermic reaction occurred in the methane-air pre-ignition period. The distance traveled by the wave to the beginning of the methane-air combustion front varies from about 3 to 5 mm, this is about one-half the distance of corresponding zones as measured by Ashman.

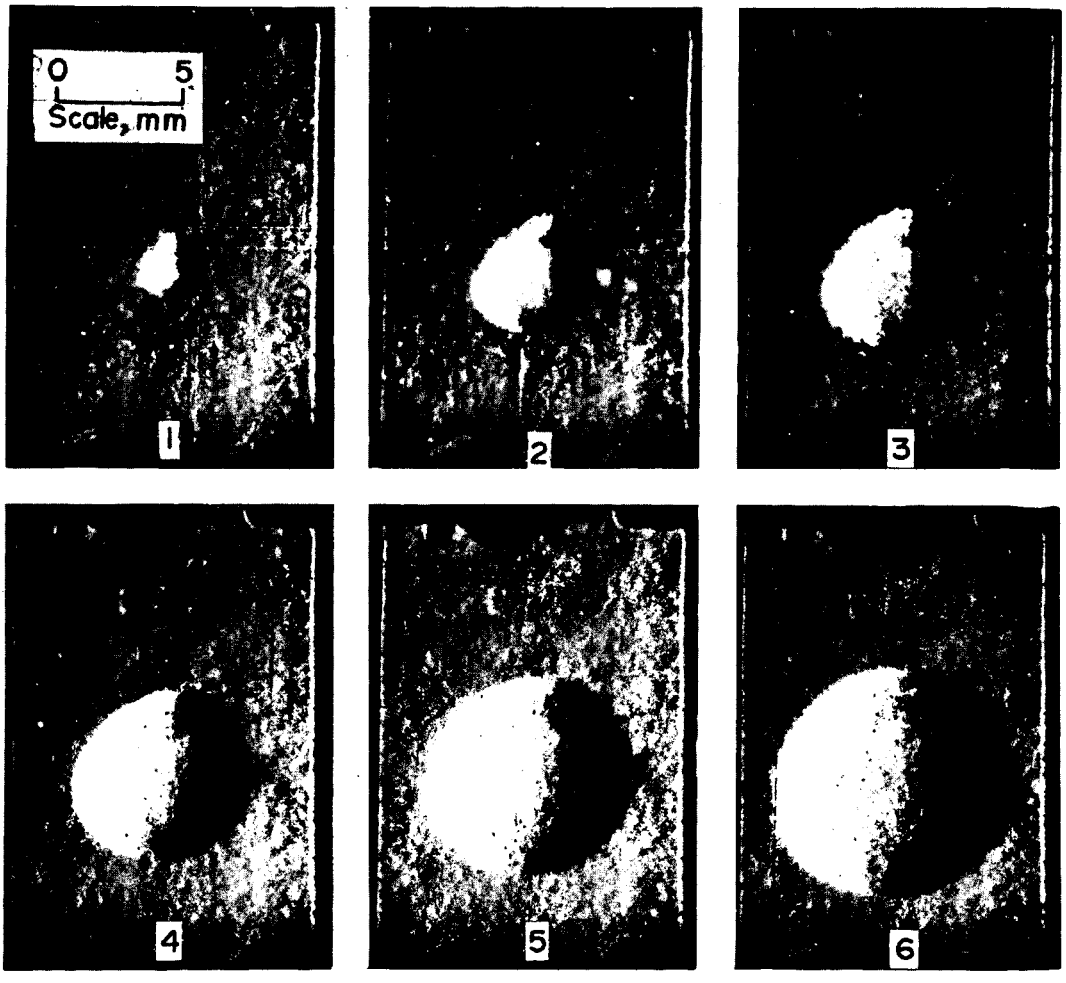

Fig. 8. - Schlieren record of laser irradiated 60 micron particle in stoichiometric methane-air mixture, 0.27 msec between frames.

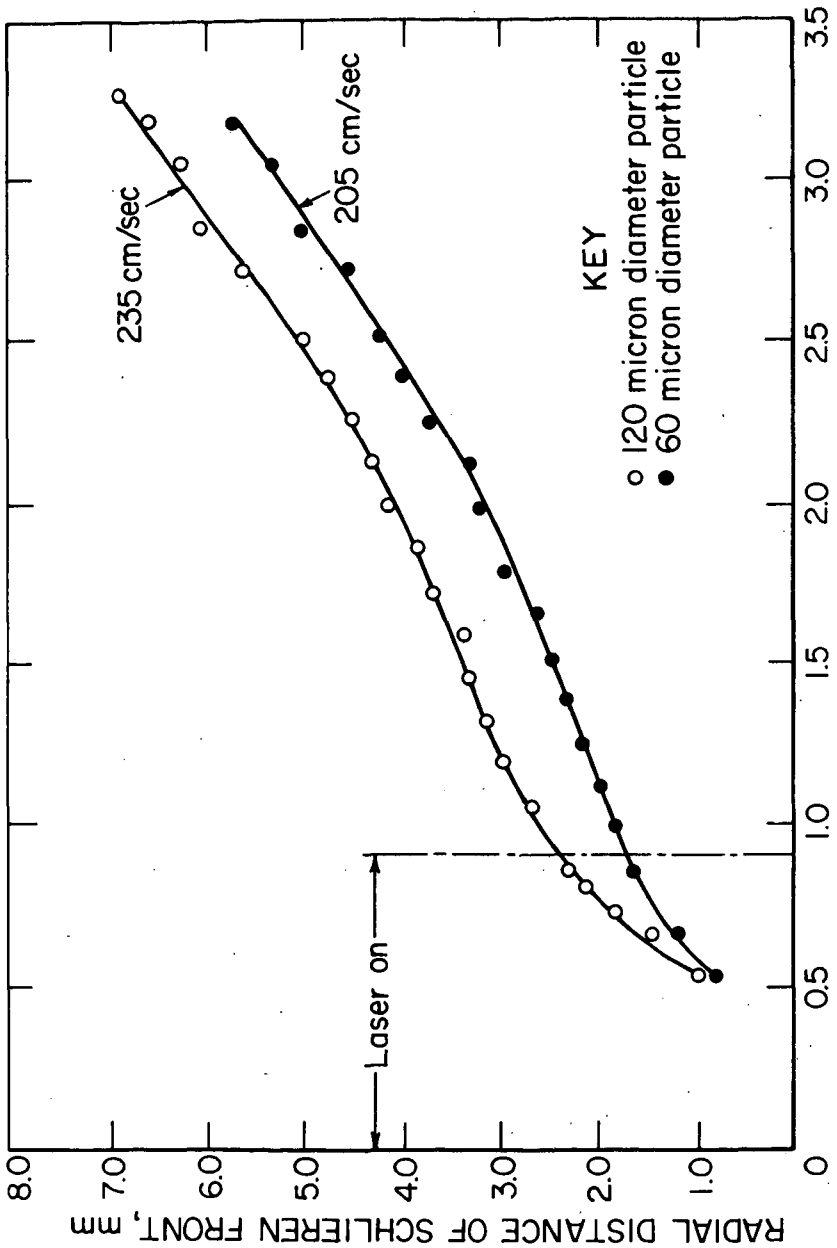

Fig. 9.- Schlieren growth surrounding a single magnesium particle irradiated by critical laser power density to ignite stoichiometric methane-air mixture.

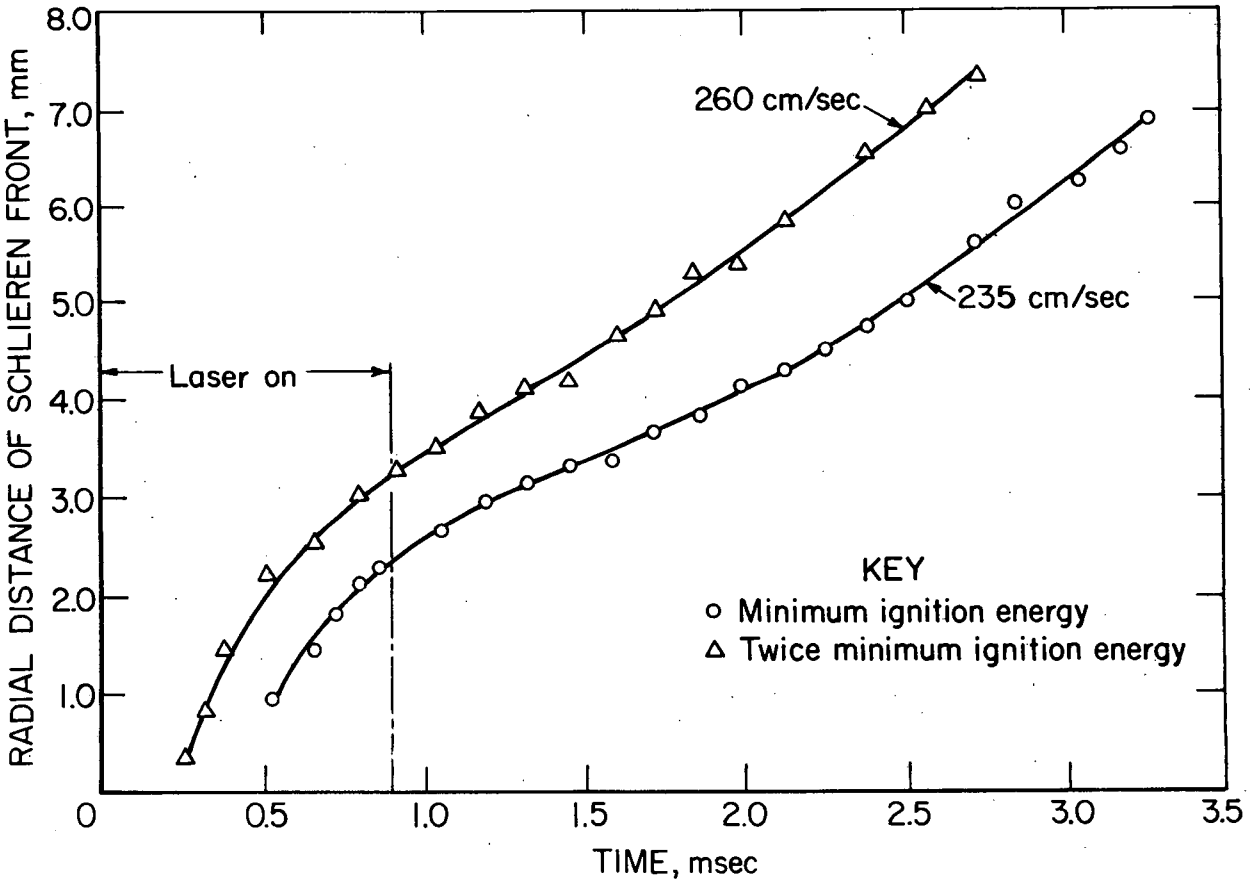

Fig. 10. - Schlieren growth surrounding a single 120 micron diameter magnesium particle during laser irradiation and ignition of stoichiometric methane-air mixture.

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